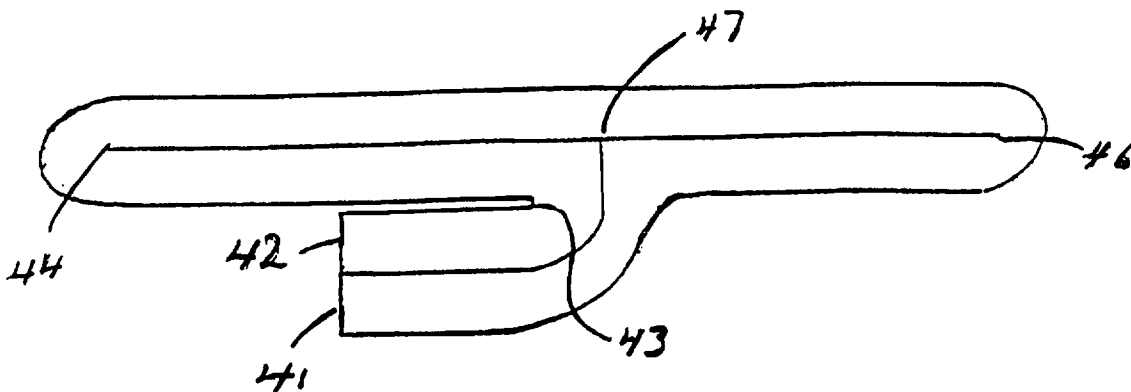




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(54) Title: HERMETICALLY SEALABLE FILM AND METHOD OF MAKING



(57) Abstract

The present invention is a multi-layer film which has an improved composite structure for providing hermetic seals to packages manufactured in high speed packaging apparatus. The structure of the multi-layer film of the present invention includes a main substrate and a sealant layer. The sealant layer, in turn, includes two components, an intermediate layer which has the primary function of compliance during sealing, and a sealing layer which has the primary function of providing adhesivity to the completed seal. As a result of this invention, high strength hermetic seals can be provided to multi-layer films effectively and efficiently, and hermetic seals are provided with the ability to be peeled open without destruction of several layers of the film.

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## HERMETICALLY SEALABLE FILM AND METHOD OF MAKING

The present invention relates to the art of packaging using multi-layer films, and, in particular, to a new composite multi-layer film for providing hermetic seals to multi-layer film packages.

5 In certain cases, plastic films can be prepared which exclude moisture and oxygen, but permit the passage of light. In other cases, it is also important to prevent light from passing through the film barrier. Barrier properties can also be modified and/or enhanced by treatments such as heat and flame treatment, electrostatic discharge, chemical treatments, halogen treatment, -violet light, and combinations thereof.

10 A primary concern for designing multiple-layer plastic films for packaging is to ensure they can be processed on high speed form/fill/seal machinery. Form/fill/seal package apparatus operates by unwinding continuous film from bulk film rolls, followed by forming pouches therefrom, filling the pouches, and, finally, sealing the pouch closed. Thus, the film must have sufficient flexibility to undergo machine folding from a flat orientation to a folded condition, and be subjected to a sealing function which is part of high-speed packaging apparatus. In selecting the optimum multi-layer film for its barrier properties, high-speed unrolling and folding are the primary concern. An additional, and very important aspect of the packaging process, however, is the ability to effectively seal the pouch after it is filled with the product.

20 Today, horizontal form/fill/seal ("HFFS") and vertical form/fill/seal ("VFFS") apparatus include sealing functions at various stages of the packaging process and at high speeds. In an HFFS apparatus, individual pouches are formed by folding the multi-layer film in half followed by providing vertical seals along the length of the folded web and separating the pouches along the seals formed by vertical sealing. (Optionally, the bottoms of the pouches can also be sealed). After the formed pouch is filled, the top of the pouch is sealed.

25 Similarly, in VFFS apparatus, the continuous web is formed around a tube and the web is immediately joined together by a longitudinal sealing jaw as either a lap seal or a fin seal. A lap seal is depicted schematically in Figures 1 and 1a. A fin seal is depicted schematically in Figures 2 and 2a.

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A second sealing function is present in a VFFS configuration which consists of a combination top- and bottom-sealing section (with a bag cut-off device in between). The top-sealing portion seals the bottom of an empty bag suspended from the bag forming tube while the bottom portion seals the top of a filled bag.

5 As a consequence of processing high-barrier property multi-layer films in high speed form/fill/seal apparatus, a structural phenomenon known as "tunnels" are formed. Tunnels are formed at the overlap of film which occurs when the film is folded, such as at the bottom of pouches formed on HFFS apparatus and at the sides of pouches formed on VFFS apparatus. Tunnels are also formed at the convergence of film web at a fin seal  
10 area. The examples of tunnels 34, 36, and 37 are depicted in Figure 3.

In order, therefore, to provide high-barrier multi-layer film with hermetic seals, several factors must be considered. It is important to provide a sealing capability at as low a temperature as possible in order to retain, among other things, stereoregularity imposed during orientation, little or no film shrinkage, retention of film and/or chemical  
15 additive properties, and highly consistent quality sealing capabilities. Furthermore, the film must have surface characteristics which permit it to be readily used on high-speed machinery. For example, the coefficient of friction must be such that it can be readily unrolled from a high volume roll of film and passed through the packaging machinery. Undesirable sticking or friction characteristics can cause bag imperfections and  
20 interruption of high-speed processing. Moreover, seals formed during process must have good seal strength.

Additionally, it is also desirable to provide seals which can be readily opened by the consumer without creating a "z-direction" tear. A z-direction tear is one which causes the disruption of the integrity of the multi-layer film when the film is pulled apart  
25 at the seal. Figure 8 depicts a z-direction tear. A z-direction tear is one which does not simply separate at the line at which the seal is formed. Instead, the separation extends to layers of the film which ripped along the layers thereof and not simply delaminated one from another. As a result of z-direction tears, it is difficult to reclose such packages to maintain freshness of the contents. When z-direction tears can be eliminated, packages are easily refolded and sealed by a mechanical means such as a clip imposed over the  
30 folds of the once-opened package. Z-direction tears can result when the adhesive

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properties used to seal the package create a tensile strength greater than that required to disrupt the integrity of one or more of the layers of the film web. It is desirable to produce a package which will separate along the seal during opening so that the package can be easily reclosed and secured against passage of air.

5           The present invention provides the packaging artisan with the ability to improve total package barrier by eliminating "tunnels" which can be formed at the overlap of film such as at fold locations and at the center lap of a fin seal area. As a result of the present invention, multi-layer films which are used in packaging can be provided with excellent sealing capability without the necessity of expensive sealants or a thick sealant layer  
10           which causes difficulty on high speed packaging machines. Furthermore, the present invention eliminates the need to load silicone in an outer layer or lamination of a multi-layer film structure to facilitate passage through packaging apparatus. The present invention also eliminates inconsistent packaging machine performance. The present invention also significantly increases seal strength without reliance on a super-adhesive  
15           which may not be compatible with high speed processing. In addition to the advantages set forth above, the present invention has also improved packaging prepared in accordance with the invention by eliminating z-direction tear upon separation of the sealed films. Thus, peelable hermetic seals can be provided.

20           Figure 1 is a schematic representation of a flexible multi-layered film package formed on a vertical form/fill/seal apparatus and joined by a lap seal;

            Figure 1a is a schematic cross-section taken along line a-a of the package depicted in Figure 1 wherein the lap portion of the seal is exploded for illustration;

            Figure 2 is a schematic representation of a package formed on a VFFS apparatus which includes a fin seal closure;

25           Figure 2a is a cross-section of the package shown in Figure 2 taken along line a-a wherein the fin seal is exploded for illustration;

            Figure 3 is an oversized schematic of the seal formed along the bottom of Figure 2 and shown from the view provided from line 3-3;

30           Figure 4 is a schematic representation of the view depicted in Figure 3 in which a seal has been made of a film prepared in accordance with the present invention;

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Figure 5 is an exaggerated cross-section of a multi-layer film prepared in accordance with the present invention;

Figure 6 is also a schematic representation of a cross-section of a preferred film formed in accordance with the present invention;

5 Figure 7 is another schematic representation of a cross-section of a preferred film formed in accordance with the present invention;

Figure 8 is a schematic representation of a multi-layer film which exhibits z-direction tear; and

10 Figure 9 is a cross-section of a portion of a seal jaw which depicts the shape and dimension of seal jaw teeth.

The present invention is a multi-layer film for forming hermetic seals on packages and a method of improving the seal characteristics of multi-layer films which are hermetically sealable in high-speed packaging machines. In order to provide a hermetic seal to packages formed from multi-layer films, care must be taken to provide a sealing  
15 medium which accommodates the nature of the barrier film used for the package, i.e., its modulus or stiffness, thickness, adversity to temperature and pressure imposed under sealing conditions, etc. "Hermetic seals" as used herein means both peelable and unpeelable seals which provide hermetic barrier properties, i.e., does not permit passage of a gas such as air.

20 "Multi-layer film" as used in the present application means a film having more than one layer of material which forms the final film product. For example, while it is known that oriented polypropylene (OPP) is excellent for use in packaging, in order to provide additional barrier properties, it is also known to overlay oriented polypropylene with additional layers of polymers and/or other additives. Such films can be prepared by  
25 coextrusion of two or more layers simultaneously, or by extruding subsequent layers in series, or by coating, laminating, or combinations thereof. Multi-layer films of the present invention includes the entire web structure as it is used for packaging.

The multi-layer films for forming hermetic seals on packages of the present invention comprises two components, (a) a main film substrate and (b) a sealant layer  
30 adjacent and bonded to the main film substrate comprising (I) an intermediate layer bonded to said main film substrate and having sufficient flow property under sealing

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conditions to deform and comply with all unfilled space between sealing jaws during sealing; and (ii) a sealing layer bonded to said intermediate layer to provide a seal under sealing conditions; and (c) a composite structure of said main film and said sealant layer such that said intermediate layer has a composition different than said sealing layer's composition, and said sealant layer has sufficient thickness in relationship to the thickness of said main film substrate to eliminate tunnels being formed during sealing whereby hermetic seals are formed during sealing.

The inventors have discovered that when a main film substrate is provided with a sealant layer having two separate components, each primarily designed to fulfill one of the required sealing functions, imperfections in hermetic seals normally associated with high-speed film packaging can be effectively and efficiently eliminated. Specifically, a "sealant layer" is provided to a main film substrate. The sealant layer has two components, an intermediate layer and a sealing layer. The "intermediate layer" primarily meets the requirement of "compliance" throughout the volume between the surfaces of sealing jaws of high-speed packaging apparatus during the sealing function. The "sealing layer", on the other hand, primarily meets the requirement of providing high performance adhesion under sealing conditions. Bearing in mind that sealing conditions include both high temperature and pressure imposed on the sealant layer, both the intermediate layer and the sealing layer will participate in both of the sealing functions, i.e., compliance and adhesion. However, the primary function of the intermediate layer is to provide compliance while the primary responsibility of the sealing layer is to provide adhesivity. Thus, the composition of the intermediate layer is different from the composition of the sealing layer.

The sealant layer is provided to the main film substrate such that the intermediate layer is adjacent to and bonded directly to the main film substrate while the sealing layer overlays and is the outermost skin of the sealant layer.

"Compliance" in the context of the present invention means the ability to be easily and non-elastically deformed to fill and conform to the entire space between the sealing surfaces of a sealing jaw. Sealing jaws can operate from a temperature of from about 120°C to about 190°C, and normally are imposed on a film packaging material at a pressure of from about 120 psi to about 180 psi (about 827.4 kPa to about 1241.1 kPa).

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Sealing jaws can be flat, or, in many cases, are provided with teeth. In Fig. 9, a section of a sealing jaw 90 having teeth 94 is schematically depicted in perspective. The jaw 90 is formed with teeth 94 having tooth width  $W$ . A complementary jaw is used in conjunction with jaw 90 so that the peak of the teeth mesh with the valleys 93 of the jaw. The surfaces of the jaws close in the sealing position on two multi-layer films, thereby clamping the films therebetween. To form a hermetic seal, the volume between the surfaces must be completely filled during sealing. These are the normal sealing conditions under which the intermediate layer must be capable of compliance.

The intermediate layer must have sufficient material to undergo compliance without leaving a void. The intermediate layer must be sufficiently thick such that a continuum of material is provided throughout the space between the surfaces of the sealing jaw. The flow property of the intermediate layer must be such that in the presence of the temperature and pressure exerted during sealing, the material maintains a viscosity which is easily deformed but maintains a non-interrupted mass throughout the space between the sealing surfaces.

Polyethylene, and its co- and terpolymers are contemplated for use in the intermediate layer. Linear low density polyethylene (LLDPE) has been found to be an excellent component for the intermediate layer. Linear low density polyethylene is inexpensive and has the correct compliance requirements for the intermediate layer. Linear low density polyethylene used in the intermediate layer can be used alone or in combination with other components such as a random copolymer of ethylene and propylene or a random terpolymer of ethylene-propylene-butylene (EPB). The intermediate layer can also have a component selected from the group consisting of amorphous polyamide (e.g. nylon), ionomers and mixtures thereof.

The sealing layer of the sealant layer, in turn, has the primary responsibility of providing adhesivity. Thus, sealing layer components should be selected based on their ability to provide good adhesive seal strength, i.e., adequate tensile strength of the seal. Inasmuch as the primary function of the sealing layer is that of adhesivity, the thickness of the sealing layer is less than the thickness of the intermediate layer. It has been found that polyethylene homo-, co- and terpolymers, amorphous polyamide (e.g. nylon), ionomers and mixtures thereof are useful. Specific components include ethylene-propylene random

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copolymers, ethylene-propylene-butene random terpolymers, and ethylene-butene polymers referred to as metallocene plastomers prepared by a metallocene catalyst. These can be the primary component of the sealing layer.

5 In order to enable the sealant layer to fulfill the two functions simultaneously during the act of sealing, the present invention must include a "composite structure" which establishes a relationship between the sealant layer and the main film substrate. The term "composite structure" is used herein to refer to the relationship of the thickness of the sealant layer to the thickness of the main film substrate. Basically, the thickness of the sealant layer must be sufficient in relationship to the thickness of the main film  
10 substrate to ensure compliance and total uninterrupted surface-to-surface contact of the sealing layer.

Generally, the film which is overlapped by being folded or bent back on itself forms "tunnels." "Tunnels" are the passages which remain at the folds or bends of the film even after sealing. Tunnels also occur at the convergence of multi-layer film at a fin  
15 seal area. (See tunnel 37 at Figure 3). When the multi-layer film is thinner, the tunnels formed at the bends are smaller. The thicker the multi-layer film, the greater the dimension of the tunnels formed by bends and convergence. Thus, as the thickness of the multi-layer film increases, the sealant layer thickness must also be increased to ensure compliance and surface-to-surface contingency.

20 The minimum thickness for any multi-layer film can be found by solving for  $S_0$  in the mathematical relationship set forth below wherein

$$S_0^3 = \left[ \left( \frac{2.15}{96} \right) \left( \frac{LW}{L+W} \right) \left( \frac{G'}{T} \right) \right] t^2.$$

wherein,

$S_0$  (cm) is said sealant layer thickness,

$t$  (cm) is total thickness of the composite structure,

25  $G'$  (dynes/cm<sup>2</sup> (mN/m<sup>2</sup>)) is the storage shear modulus of said sealant layer

( $G'$  is temperature and rate dependent)

$T$  (dynes/cm<sup>2</sup> (mN/m<sup>2</sup>)) is applied compression stress on a seal jaw,

$L$  (cm) is jaw seal length, and

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W (cm) is a seal tooth width.

The thickness of the composite structure, t, is the thickness found after all layers of the final film product are added. Thus, if an initial film web is formed by coextrusion of the sealant layer in combination with one or more additional layers which do not comprise the entire multi-layer film, the total thickness, t, of the composite structure cannot be calculated until the additional layers are added by extrusion, lamination, coating, or otherwise.

In most cases, it has been found that the sealant layer has a thickness of from about 15% to about 70% of the thickness of the composite structure, and is preferably in many embodiments from about 20% to about 60% of thickness of the composite structure.

Focusing on the sealant layer, it has been found that the thickness of the intermediate layer is from about 10% to about 90% of the thickness of the sealant layer, and is preferably from about 40% to about 80% of the thickness of the sealant layer.

Referring now to the drawings, the present invention is explained in relationship to lap seal and fin seal packages. Figure 1 is a schematic representation of a package formed by closing a multi-layer film by a lap seal. A multi-layer film web has been closed to form the package 10 by joining web edges 11 and 12 at an overlap 13. Figure 1a is a cross-section schematic of a view taken from line a-a in Figure 1 which shows the overlapping relationship of the edges 11 and 12. The package is also depicted schematically as being sealed at either end by top seal 15 and bottom seal 16. This type of package is formed on a VFFS apparatus and tunnels will be formed in the ends 16' and 17' of seals 15 and 16.

Similarly, Figure 2 and 2a depict a package formed on a VFFS machine by the formation of a fin seal 25. The fin seal 25 is formed by web edges 21 and 22 together and bending the joined ends back towards the package at bend 23 to form the fin seal 25. The package is also depicted as having end seals 26 and 27 at the top and bottom thereof, respectively.

In order to clearly depict the phenomenon of tunnels formed during sealing, reference is made to Figure 3 which is an oversized end view of the fin seal package shown in Figure 2 and taken along lines 3-3. From this end view, the fin seal 35 can be

seen as formed by joining edges 31 and 32 and bending the joined portion thereof at bend 33. As the end is sealed, the thickness dimension and stiffness of the film create tunnels 34 and 36 at each edge of the package, while yet another tunnel 37 is formed at the convergence of the multi-layer film at the fin seal area.

5 Preparation of a package in accordance with the present invention eliminates the tunnels normally associated with sealing the package ends of continuous high speed packaging systems. In Figure 4, a package prepared in accordance with the present invention is schematically depicted with a view similar to that shown in Figure 3. Specifically, the edges of a multi-layer film 41 and 42 have been joined and lapped back at bend 43 to form a seal fin. Tunnels usually formed as a result of sealing the end of a  
10 fin seal package have been eliminated. Thus, the tunnels found at ends of the seal found at side bends 44 and 46 have been completely closed. Similarly, the convergence 47 of the multi-layer film to form the fin seal shows the elimination of the tunnel depicted at convergence 37 in Figure 3.

15 Referring to Figure 5, the structural relationship of films prepared according to the present invention in its most basic aspect is schematically depicted. The composite structure 52 is shown as having a thickness  $t$ . The composite structure includes a relationship between the sealant layer 54 and the main film substrate layer 56 which is sufficient to ensure both i) compliance by operation of intermediate layer 53, and  
20 ii) sealing by sealing layer 55. The main film substrate is simply depicted as having at least one layer 58 and possibly an additional layer 57. In its simplest manifestation, the overall composite structure 52 can include, for example, an oriented polypropylene which makes up the entire main film substrate 56. Optionally, an additional high density polyethylene (HDPE) can be provided as a layer 57. The intermediate layer 53 can also  
25 be, for example, a single component, such as linear low density polyethylene, and a sealing layer 55 can be composed of an ethylene-propylene-butene EPB random terpolymer as the sole component.

The main film substrate of the present invention can be those single or multiple-layer films used in the art of packaging in order to provide simple or enhanced barrier  
30 properties for maintenance of package contents. For example, the substrate can include one or more layers selected from the group consisting of oriented polypropylene,

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polypropylene, ethylene-propylene copolymers, polyethylene terephthalate, polyamide, polyacrylonitrile copolymer, polyvinylidene chloride, fluoro-polymers, ethyl-vinyl alcohol copolymers, and mixtures thereof. Other layers can be barrier resins, tie resins, metallized film, ceramic deposited film (e.g., SiO<sub>4</sub>), plasma chemical vapor deposited film, and metal, ceramic, plasma chemical vapor. For example, the main film substrate can include oriented polypropylene, and additional layers, such as high density polyethylene, can be coextruded with the oriented polypropylene. Layers which provide additional moisture, oxygen, and light barrier properties, can also be included in the main film substrate.

A high barrier film can be provided which includes a substrate made of a propylene homopolymer or copolymer, especially oriented polypropylene, which has a surface which also includes a maleic acid anhydride modified propylene homopolymer or copolymer. At least one surface has a skin layer of ethylene vinyl alcohol copolymer.

For a main film substrate layer (e.g. layer 56 of Fig. 5) which employs ethylene vinyl alcohol, films contemplated are of the kind described in U.S. Patent Nos. 5,153,074 to Migliorini et al.; 4,704,954 and 4,852,752 both to Duncan; 4,347,337 to Knott, II; 5,192,620 to Chu et al.; 5,221,566 to Tokoh et al. And Japanese Kokai No. 1 [1989]-267032.

Figure 6 depicts a composite structure 60 which includes a sealant layer 62 and a main film substrate 66. The sealant layer 62 includes a sealing layer 61 which is a 5 gauge (1.27 microns) layer of an ethylene-propylene-butene random copolymer sold under the trade name Chisso 7701 by the Chisso Chemical Company of Japan. The intermediate layer 63 is a 20 gauge (5 microns) layer which includes a combination of a linear low density polyethylene LL3001 sold by Exxon, and the terpolymer Chisso 7701 terpolymer in a ratio of 70:30 LLDPE to EPB copolymer.

The main film substrate 66 is a high barrier film which includes a 62 gauge (15.9 microns) thick oriented polypropylene layer 65 provided as polypropylene homopolymer Fina 3371 by Fina Oil Company. Immediately adjacent the oriented polypropylene is provided a "tie" resin layer 67. The primary component of the tie layer is an anhydride grafted polypropylene provided by Mitsui under trade name Admer QF500A. The anhydride grafted polypropylene has a melt flow of 3.0 grams per ten minutes and a

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density of 0.90. The melting point,  $T_m$  is 160°C. The tie resin layer is 10 gauge (2.54 microns).

Finally, a barrier layer 68 is provided at a thickness of 3 gauge (0.8 microns). In this embodiment, the additional barrier layer is ethylene-vinyl alcohol copolymer which includes 48% ethylene. The primary ingredient of this layer is ethylene-vinyl alcohol copolymer having a melt index of 14.0, a density of 1.12 and a melt temperature of 158°C and is sold under trade designation EC-G156 by Eval.

Other barrier resins can include polyvinyl alcohols, polyvinylidene chloride, nylon, liquid crystal polymers, and fluoropolymers.

The sealant layer is 25% of the thickness of the overall composite. The intermediate layer, in turn, is 80% of the thickness of the sealant layer.

Another example of a high barrier film with which the present invention can be used is described once again in reference to Figure 6. In this embodiment, the sealing layer 61 is of the same material and same gauge as in the previous example. The intermediate layer 63 has the same composition, but is a 35 gauge (8.9 microns) layer rather than a 20 gauge (5 microns) layer. The oriented polypropylene layer 65 has the same composition as in the previous example, but is of 47 gauge (12microns) rather than 62 gauge (15.7 microns). The tie layer 67 and the additional barrier layer 68 have the same composition and dimension as in the previous example. In this second preferred embodiment, the relationship of the thickness of the sealant layer to the composite thickness is 40%. The intermediate layer forms 87.5% of the thickness of the sealant layer.

Another embodiment of the present invention includes the use of metallized high barrier films. This embodiment is explained by reference to Figure 7. The sealant layer 72 includes a sealing layer 71 and an intermediate layer 73. The sealing layer and the intermediate layer can have the same properties and be in the same proportion and having the same dimension as described above in the previous examples.

The metallized high barrier main film substrate can include a polypropylene homopolymer core 75 followed by a tie layer 77, another barrier layer 78 and a metal layer 79 which can be of the same or similar materials and have the same dimensions as described above in the previous high barrier examples. The metal layer 79 can be

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aluminum which readily adheres to ethylene-vinyl alcohol copolymer such as the layer 78, which has been described in the previous high barrier embodiments.

The composite structure 70 has the same structural relationship of thickness as described in the previous two examples. The metal adhesion assumes virtually no  
5 additional thickness since it adheres to and becomes part of the EVOH layer. The optical density achieved using this metallized high barrier film with the present structure is 2.3.

In the examples set forth herein, which are actual examples, reference is made to Figure 5 where the sealant layer 54 is composed of a sealing layer 55 and an intermediate layer 53. The main film substrate 56 can include one layer 58 or an additional layer 57.

#### 10 EXAMPLE 1

A multi-layer film was prepared in accordance with the present invention which included a main film substrate 56 having an oriented polypropylene layer 58 prepared using polypropylene homopolymer Fina 3371 from the Fina Oil Corporation. The OPP layer has a thickness of 45 gauge (11.4 microns). Immediately therebelow, the layer 57  
15 was provided at a thickness of 3 gauge (0.76 microns) using a high density polyethylene M6211 from Oxychem.

The intermediate layer 53 was provided as a 25 gauge (6.5 microns) thickness of a combination of linear low density polyethylene, LL3001 from Exxon, and EPB terpolymer, Chisso 7510, at a 70:30. The high density polyethylene, polypropylene, and  
20 the linear low density/EPB blend were coextruded.

The coextrusion was oriented in the machine direction followed by extrusion of a sealing layer 55 of EPB random copolymer, Chisso 7701 provided by the Chisso Chemical Company of Japan. This layer was provided in a 10 to 20 gauge (2.5 to 5.1 microns) dimension. The total structure was then oriented in the transverse direction in  
25 an oven to provide biaxial orientation.

Continuous samples were run at different thicknesses of sealing layer to provide a composite thickness of between 83 (21.1 microns) and 93 gauge (23.6 microns). Consequently, the composite structure provided a thickness relationship of sealant layer to main film substrate of from 42.2% to about 48.4%, while the intermediate layer ranged  
30 from about 56% to about 71% of the sealant layer.

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This example provided both high adhesivity as demonstrated by significantly improved seal strength, and fin seal packages were prepared and sealed without the presence of tunnels.

#### EXAMPLE 2

5 This example follows Example 1 except the sealing layer is an ethylene-propylene random copolymer Fina Z9470HB provided by Fina Oil Company. Samples were prepared once again at a varying thickness level of from 10 (2.54 microns) to 20 gauges (5.1 microns). The composite structure of the film prepared from Example 2 had the same range of thickness relationships as those prepared in Example 1.

#### EXAMPLE 3

10 This example follows that of Example 1 except that the sealing layer was prepared with a metallocene plastomer SLP9045 provided by Exxon. Metallocene plastomer is a ethylene-butene polymer system which is prepared with a metallocene catalyst. The composite structure of the film prepared in accordance with Example 3 had the same ratio of thickness relationships as those set forth in Examples 1 and 2.

#### EXAMPLE 4

15 This example follows Example 1 except for the sealing layer which was prepared using a polyethylene vinyl-acetate sold under the trade name Elvax 3130SB and is provided by DuPont. The composite structure contained the same range of thickness relationships as in the previous examples.

#### EXAMPLE 5

20 Example 5 follows Example 1. The intermediate layer 53, however, was made of strictly an EPB random copolymer, Chisso 7701 provided by the Chisso Chemical Company. This layer was provided at a thickness of 25 gauge (6.4 microns).  
25 Additionally, the sealing layer was SLP9045 metallocene plastomer provided by Exxon rather than EPB random copolymer. The composite structure of the film prepared in Example 5 had the same range of relationships as did those set forth in Examples 1-4.

#### EXAMPLE 6

30 In this example, a film structure was prepared as in Example 5 except that the sealing layer composition was a blend of metallocene plastomer SLP9045 and EPB random copolymer Chisso 7701 at a ratio of 50:50.

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EXAMPLE 7

This example follows Example 1 except the intermediate layer 53 was prepared from a combination of linear low density polyethylene, LL3001, and ethylene-propylene random copolymer, Fina 8573 provided by the Fina Oil Corporation, at a ratio of 70:30.

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EXAMPLE 8

This Example follows Example 7 except the intermediate layer was a blend of LLDPE, LL3001, and a metallocene plastomer, EXACT 3024, provided by Exxon, in a ratio of 80:20.

EXAMPLE 9

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Example 9 was prepared differently from the previous examples in that it contained but one layer 58 of polypropylene, Fina 3371, at a thickness of 45 gauge (111.4 microns). The intermediate layer 53 was provided at a thickness of 25 gauge (6.35 microns) and was the linear low density polyethylene, LL3001. The same EPB random terpolymer Chisso 7701 at a thickness range of 10 to 20 gauge (2.5 to 5 microns) was used. Unlike the previous examples, this entire structure was extruded together and oriented in both a machine direction and the transverse direction to provide a composite biaxially oriented multi-layer film. Samples were prepared using the range of gauges of sealing layer so that the composite structure provided a relationship of thicknesses of sealant layer to main film substrate of from about 44% to about 50%. The intermediate layer, in turn, was present in a range of relative thickness to the sealant layer of from about 55% to about 71%.

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EXAMPLE 10

Example 10 was prepared similarly to Example 9 except that the intermediate layer was changed from a single linear low density polyethylene component to a combination of LLDPE, LL3001, to low density polyethylene, LDPE, Chevron 1017, at a 70:30 ratio.

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The films prepared from Examples 1-10 can also be metallized on the high density polyethylene side. Thus, an additional metal layer can be imposed on the multi-layer film.

One or more webs can be laminated to the multi-layer films to form a new multi-layer film having a different composite structure thickness. Such webs include, but are not limited to, oriented polypropylene, polyethylene terphthalate (PET), polyamide such as

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nylon, polyethylene, and can be a mono-layer or another multi-layer film. Typically, the web forms an outer surface. It is important, however, to consider the thickness of the end product film in order to calculate the requisite thickness of the sealant layer.

### EXAMPLES OF PREPARED SEALS

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#### EXAMPLE 11

Seals were prepared at a seal temperature of 150°C and at a packaging speed of 60 packages per minute. The seal jaw frequency of impact was 2.959 per second. The seal jaw pressure was 150 psi (1034 kPa). The ultimate laminate thickness,  $t$ , of the composite structure was 195 gauge (50 microns). The width of the seal tooth,  $W$ , was 10 0.1 cm. The dimension  $L$ , one half of top or bottom seal length of the package in centimeters was 6.35 cm.

A composite sealant layer was provided using LL 3001/Chisso 7510, at a 70:30 ratio, as the intermediate layer and Chisso 7701 as the sealing layer. The  $G'$  for the composite sealant layer was  $1.3 \times 10^5$  dynes/cm<sup>2</sup> ( $1.3 \times 10^5$  mN/m<sup>2</sup>). The composite sealant layer of thickness was calculated to be 34.85 gauge or 0.000885 cm (8.85 15 microns) as a minimum requirement to provide an airtight seal.

Under the same packaging conditions, when  $G'$  is  $0.443 \times 10^5$  dynes/cm<sup>2</sup> (mN/m<sup>2</sup>) for metallocene SLP 9045 polymer as the intermediate layer and Chisso 7701 as the sealing layer, the required minimum thickness was calculated as 24.4 gauge or 0.00062 20 cm (6.2 microns).

#### EXAMPLE 12

This example follows Example 11 except a seal packaging speed of 30 packages per minute and seal jaw impact frequency of 1.29 per second was used. The required composite sealant thickness was calculated to be 24.9 gauge (or 0.000632 cm (6.3 25 microns)) to provide an airtight seal. The composite sealant layer was the same as Example 11. The  $G'$  for this composite sealant is  $0.47 \times 10^5$  dynes/cm<sup>2</sup> ( $0.47 \times 10^5$  mN/m<sup>2</sup>).

#### EXAMPLE 13

This example follows Example 12 except the temperature was 140°C. The laminate composite thickness,  $t$ , was 195 gauge. The sealant included a blend of LL3001 30

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and Chisso 7510 at a 70:30 ratio as a intermediate layer, and Chisso 7701 as a sealing layer and the  $G'$  for the sealant was  $0.66 \times 10^5$  dynes/cm<sup>2</sup> ( $0.66 \times 10^5$  mN/m<sup>2</sup>).

The minimum required thickness of the sealant layer was calculated to be 28.0 gauge or 0.000709 cm (7.11 microns) in order to provide a hermetic seal.

5           It should be noted that the heat seal temperature should be above the melting temperature,  $T_m$ , for the sealant layer if the sealant layer includes semi-crystal polymers. If the sealants are amorphous polymers such as copolyester or amorphous polyester for polyester film, or amorphous nylon for polyamide film, the heat seal temperature should be above the glass transition,  $T_g$ , temperatures of the polymers used in the sealant layer.

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**WHAT IS CLAIMED IS:**

1. A multi-layer film for forming hermetic seals on packages comprising:
- (a) a main film substrate;
  - (b) a sealant layer adjacent and bonded to said main film substrate, said sealant layer comprising;
    - (i) an intermediate layer bonded to said main film substrate and having sufficient flow property under sealing conditions to deform and comply with all unfilled space between sealing jaws during sealing, and
    - (ii) a sealing layer bonded to said intermediate layer to provide a seal under sealing conditions; and
  - (c) a composite structure of said main film substrate and said sealant layer such that said intermediate layer has a composition different than said sealing layer's composition, and said sealant layer has sufficient thickness in relationship to the thickness of said main film substrate to eliminate tunnels from being formed during sealing,
- whereby hermetic seals are formed during sealing.
2. A multi-layer film according to Claim 1 wherein said main film substrate is a high barrier film.
3. A multi-layer film according to Claim 2 wherein said high barrier film is metallized.
4. A multi-layer film according to Claim 1 wherein said main film substrate is a laminate comprising a film substrate and an outer web selected from the group consisting of paper, oriented polypropylene, polyethylene terephthalate, polyamide, polyethylene, mono- and multi-layer films, and combinations thereof.

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5. A multi-layer film according to Claim 1 wherein minimum thickness of said sealant layer in relationship to said main film substrate is determined by solving for  $S_0$  the following relationship:

$$S_0^3 = \left[ \left( \frac{2.15}{96} \right) \left( \frac{LW}{L+W} \right) \left( \frac{G'}{T} \right) \right] t^2.$$

wherein,

- 5  $S_0$  is said sealant layer thickness,  
 t is total thickness of said composite structure,  
 G' is the storage shear modulus of said sealant layer  
 (G' is temperature and rate dependent)  
 T is applied compression stress on a seal jaw,  
 10 L is jaw seal length, and  
 W is a seal tooth width.

6. A multi-layer film according to Claim 1 wherein said sealant layer has a thickness of from about 15% to about 70% of the thickness of said composite structure.

7. A multi-layer film according to Claim 1 wherein the thickness of said intermediate  
 15 layer is from about 10% to about 90% of the thickness of said sealant layer.

8. A multi-layer film according to Claim 1 wherein said intermediate layer comprises a component selected from the group consisting of polyethylene homo-, co-, terpolymers, low density polyethylene, amorphous nylon, an ionomer and mixtures thereof.

9. A multi-layer film according to Claim 1 wherein said sealing layer comprises a  
 20 component selected from the group consisting of polyethylene homo-, co-, and terpolymers, amorphous nylon, an ionomer, and mixtures thereof.

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10. A multi-layer film as described in any of the preceding claims which is made by a process comprising

(a) laying the sealant layer on said main film substrate, said sealant layer comprising,

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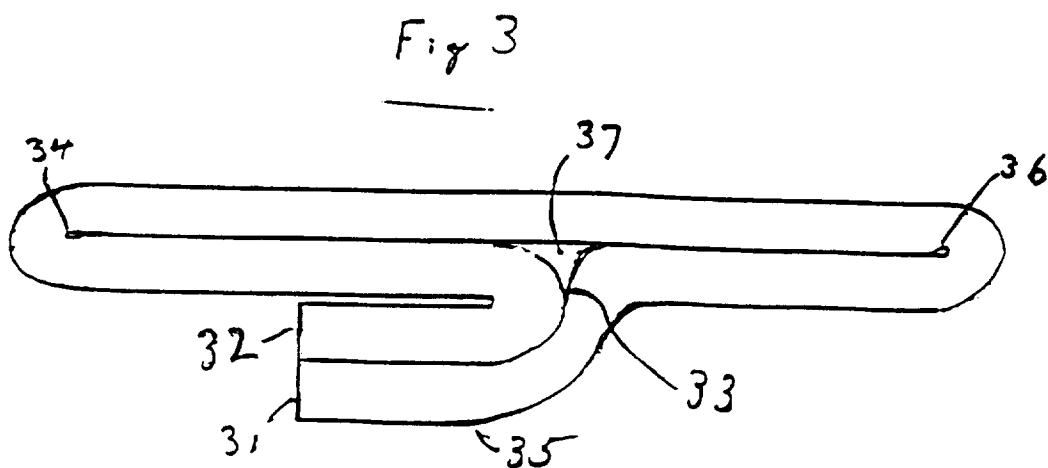
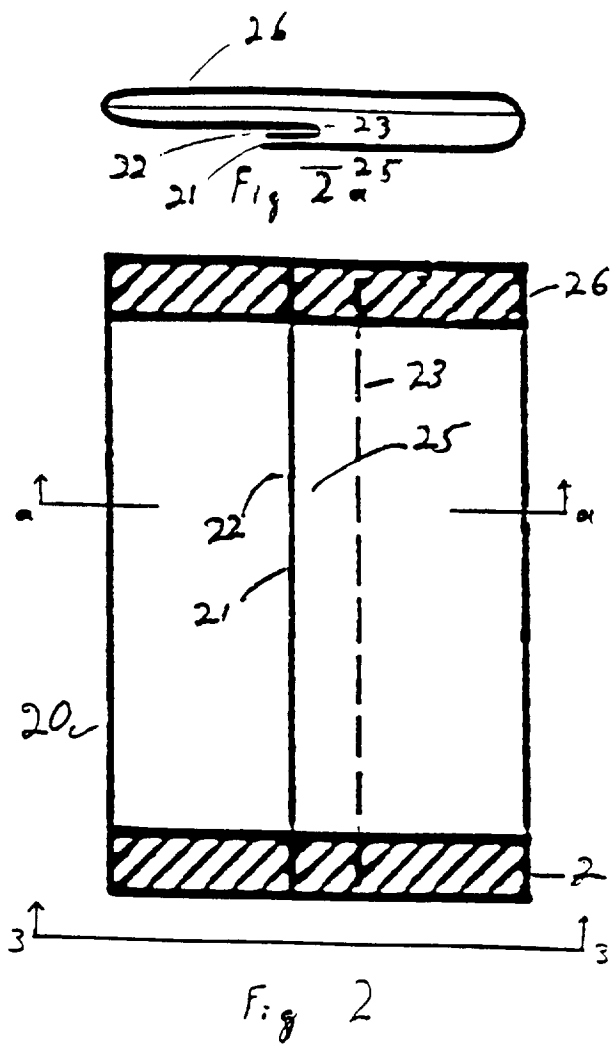
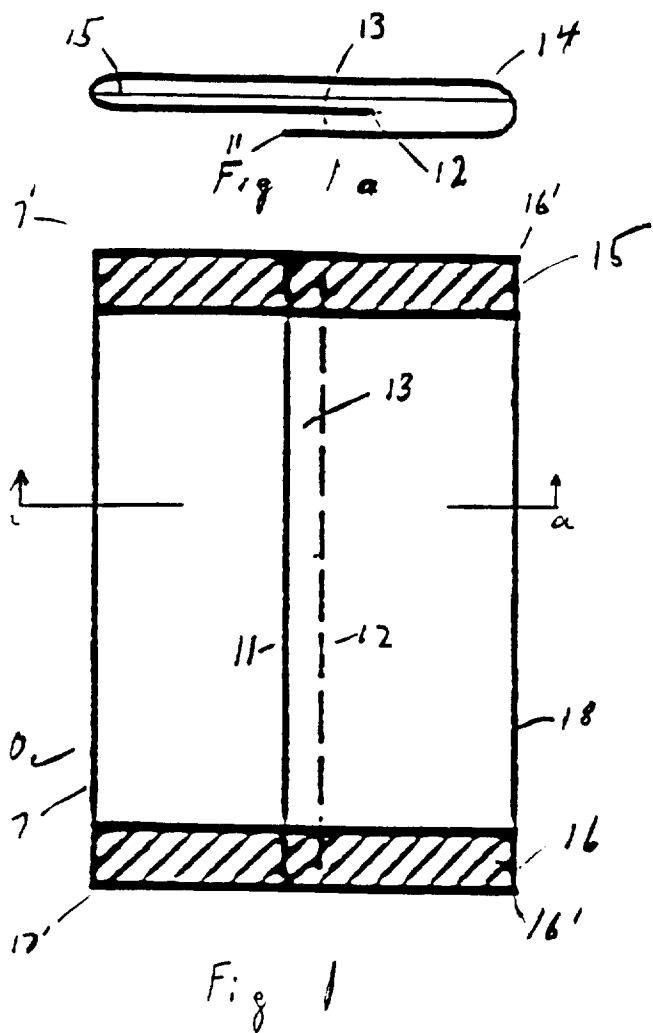
(i) an intermediate layer bonded to said main film substrate and having sufficient flow property under sealing conditions to deform and comply with all unfilled space between sealing jaws during sealing, and

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(ii) a sealing layer bonded to said intermediate layer to provide a seal under sealing conditions; and

(b) providing a composite structure of said sealant layer to said main film substrate such that said intermediate layer has a composition different than said sealing layer's composition, and said sealant layer has sufficient thickness in relationship to the thickness of said main film substrate to eliminate tunnels from being formed during sealing.

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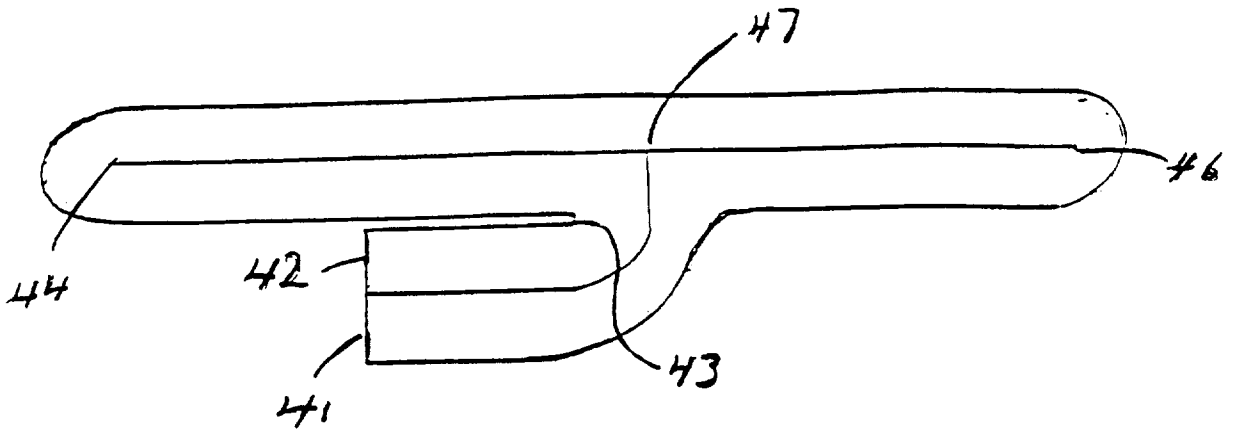


Fig. 4

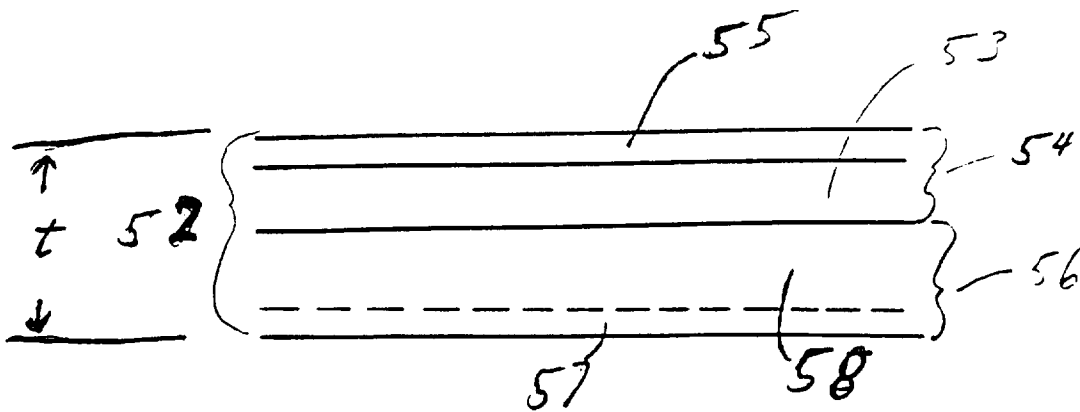
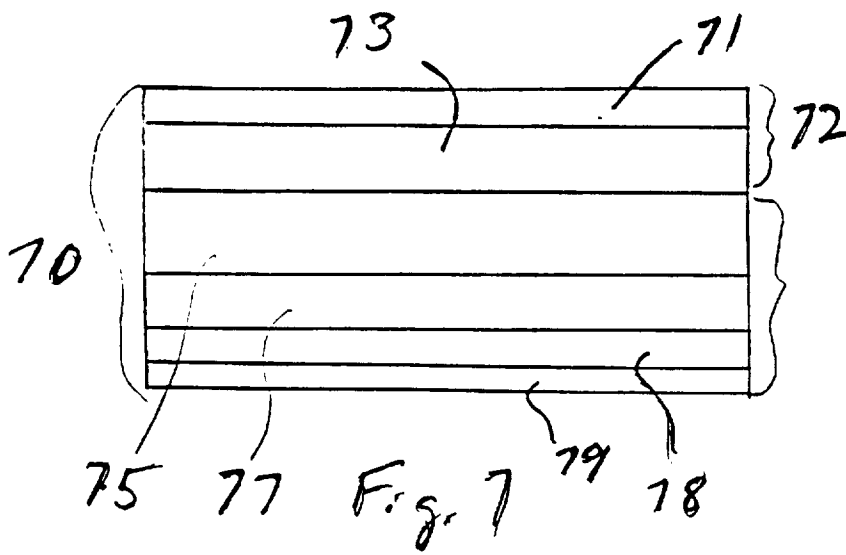
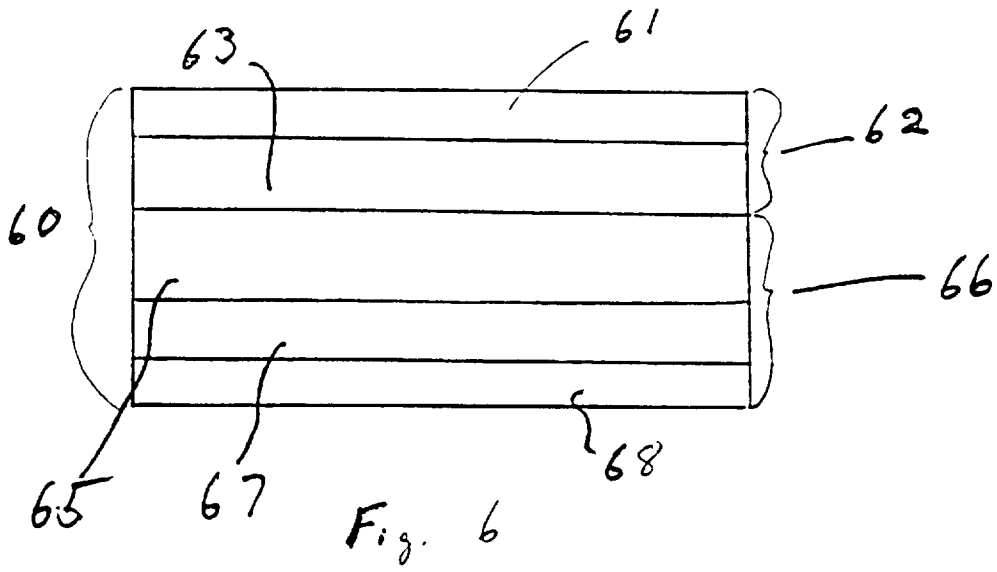


Fig. 5



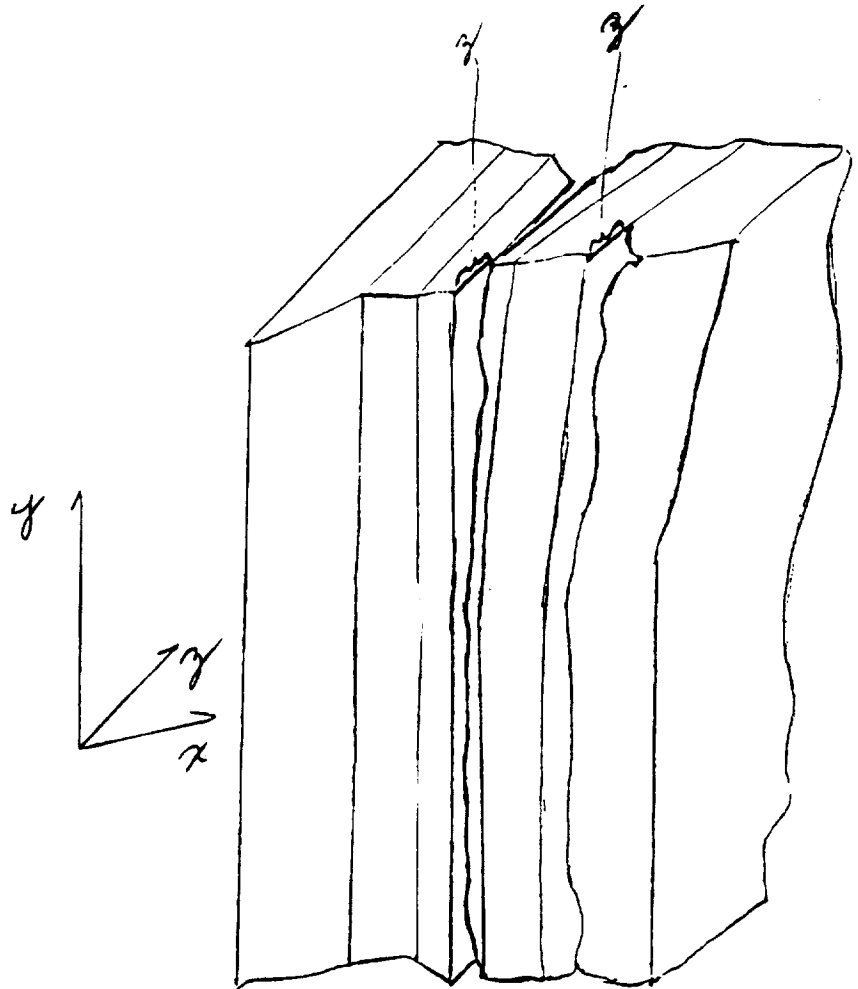


Fig. 8

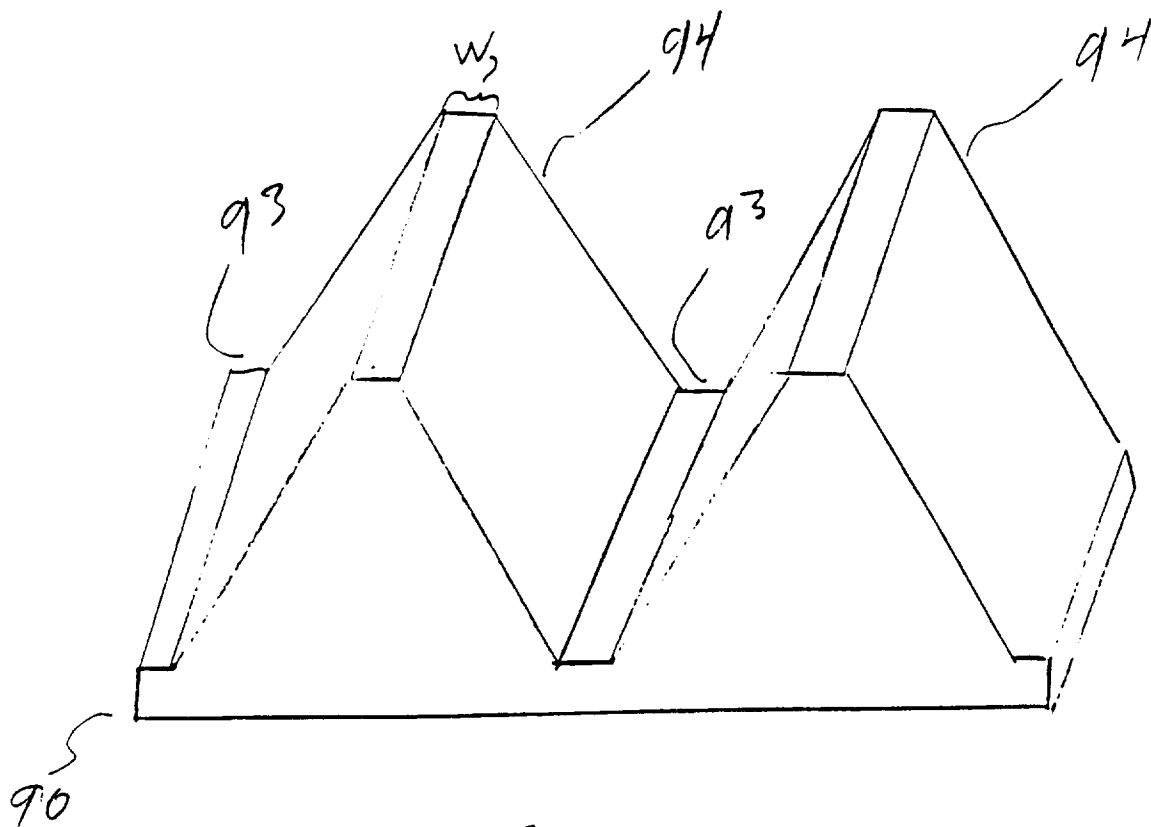


Fig. 9

## INTERNATIONAL SEARCH REPORT

International application No.  
PCT/US97/13392

## A. CLASSIFICATION OF SUBJECT MATTER

IPC(6) : Please See Extra Sheet.

US CL : 428/347, 349, 516, 517, 515, 483

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 428/347, 349, 516, 517, 515, 483

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched  
APS; search terms/package? and heat(w)seal?; hermetic?Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)  
adhesive; polypropylene; polyethylene or lldpe; Class 428, subclasses 349, 516, 517

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US, A, 4,389,450 (SCHAEFER ET AL) 21 June 1983 ( 21.06.83), Figure 1, column 3, lines 39-62, column 4, lines 6-24.	1-10
Y	US, A, 5,049,436 (MORGAN ET AL) 17 September 1991 (17.06.91) column 1, lines 11-24, column 2, lines 12-19, lines 31- 42, column 3, lines 1-11, lines 22-27	1-10
Y	US, A 4,347,337 (KNOTT, II) 31 August 1982 (31.08.82) column 1, lines 19-30	1-10
Y	US, A, 4,218,510 (WILSON) 19 August 1980 (19.08.80) column 1, lines 57-65, column 2, lines 7-10, lines 45-51, lines 55-59, column 3, lines 49-54.	1-10

 Further documents are listed in the continuation of Box C.  See patent family annex.

* Special categories of cited documents:	*T*	later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
*A* document defining the general state of the art which is not considered to be of particular relevance	*X*	document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
*B* earlier document published on or after the international filing date	*Y*	document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
*L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	*A*	document member of the same patent family
*O* document referring to an oral disclosure, use, exhibition or other means		
*P* document published prior to the international filing date but later than the priority date claimed		

Date of the actual completion of the international search

16 OCTOBER 1997

Date of mailing of the international search report

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## INTERNATIONAL SEARCH REPORT

International application No.  
PCT/US97/13392

## C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US, A, 5,346,763 (BALLONI ET AL) 13 September 1994 (12.09.94).	1-10
A	US, A, 4,384,024 (MITCHELL ET AL) 17 May 1983 (17.05.83).	1-10

**INTERNATIONAL SEARCH REPORT**

International application No.  
PCT/US97/13392

**A. CLASSIFICATION OF SUBJECT MATTER:**  
IPC (6):

**B32B 27/08, 27/32, 7/02**